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Spectroscopic and structural aspects of femtosecond dynamics of reflectivity of AlGaAs/GaAs superlattices

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Abstract. In this paper, we present results of studying dynamics of nondegenerate (two-colour) photoreflection of the AlGaAs/GaAs superlattices. These results show unique potential of these methods (i) for identification of sources of inhomogeneous broadening of optically-active 2D excitons, (ii) for analysis of fine structure of excitonic states hidden in inhomogeneous broadening, and (iii) for determination of spectral distribution of probability of radiative annihilation of 2D excitons and its relation to structural disorder.

Spectroscopy of quantum beats in 2D excitonic states

Beginning from the pioneering works [1, 2], the spectroscopy of quantum beats (SQB) has occupied a stable position in the row of other methods of the superhigh-resolution spectroscopy [3], its merits being mostly associated with its capability of revealing the structure of excited-states hidden in the inhomogeneously (Doppler- or statically) broadened bands.

For the SL, the ability of the SQB to detect hidden structure of the states can be interestingly revealed in the interference of the "bright" (B) ($J = 1$) and "dark" (D) ($J = 2$) HH-excitons. The structural defects induce an anisotropic potential of localization, which causes splitting of levels of the localized excitons [4, 5]. Thus, the spectrum of the quantum beats of a uniform ensemble of localized HH-excitons may contain up to six different frequencies. Preparation of coherent superposition of the B-and D-excitons with comparable amplitudes by direct optical excitation is impossible because of huge difference between dipole moments of the corresponding transitions. However, it can be produced by populating these states via recombination from optically excited states of the 2D (e-h)-continuum provided that this mechanism is characterized by the rates much higher than the highest frequencies of the quantum beats. The ac component of the differential reflectivity excited by this way, and the Fourier-transform (spectrum) of the quantum beats for the $SL = 30 \times (7\,\mathrm{nm}\,\mathrm{Ga}_{0.7}\mathrm{Al}_{0.3}\mathrm{As}/7\,\mathrm{nm}\,\mathrm{GaAs})$ grown by the MBE technique with no growth interruption are shown on Fig. 1.

A number of conclusions of principal importance can be drawn based on qualitative analysis of these results. First of all, interest is the fact itself that the quantum beats are detected in the subsystem of the "bright" and "dark" states. This, to the best of our knowledge, is, basically, the first direct observation of the process of quantum relaxation of states, having no alternative relaxation chanals. The second conclusion is that though the structure under study was grown with no smoothing of the hetero-interfaces, it contains a rather limited number of exciton-localizing potentials, which corresponds, as
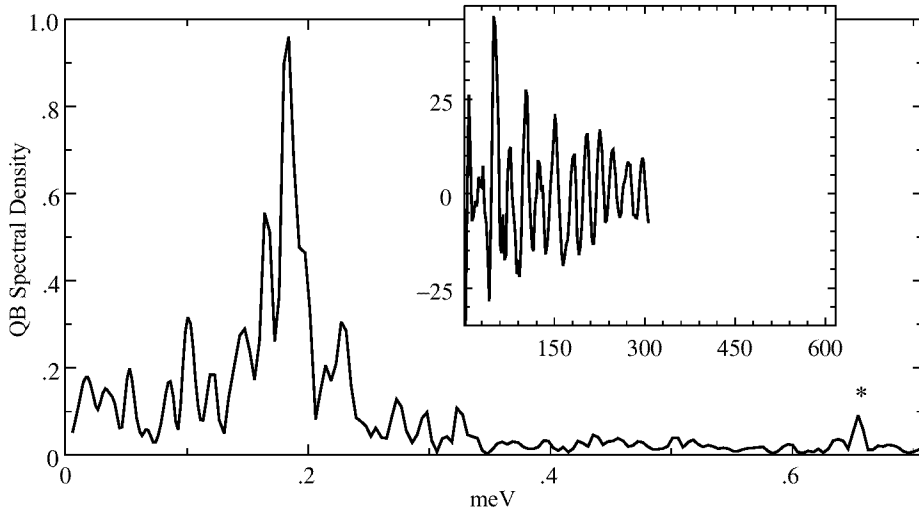
Fig 1. Ac component of the differential reflectivity and its Fourier-transform.

we belive, to few types of structural defects. The value of the symmetric exchange coupling ≈ 0.17 meV. The values of the asymmetric exchange splitting strongly depend on structure of the localizing defect and lie in the range between 0.015 and 0.08 meV. The third conclusion, which we consider to be also of great importance is that in the structures of this type the irreversible dephasing of the excitons at $T < 10$ K is determined only by the energy relaxation ($\Gamma_2 = 0.5\Gamma_1$). This conclusion is based on the fact that the quantum beats of the photoreflectivity are damping in parallel with population decay of the excitonic states (see below). And, finally, one should pay attention to the fact that the first mode of SL splitting (0.65 meV) of HH-excitons is noticable in QB spectrum. It means that rate of phonon assistant transition with this energy is less than radiative relaxation rate of localized HH-excitons ($\sim 3 \cdot 10^9 \sec^{-1}$).

Distribution of the radiative transition rates and coherence radii of HH-excitons in the GaAlAs/GaAs superlattices

For ideal GaAlAs/GaAs SL-structures, the radiative relaxation rates are expected to lie in the range of $10^{11} - 10^{12} \sec^{-1}$ [6, 7]. In real structures, the elastic scattering of 2D-excitons by impurities, fluctuations of composition, and nonuniformities of hetero-interfaces leads to smearing of low-frequency edge of the excitonic states density and confines their spatial coherence. In this case, the oscillator strengths of polaritons, which would concentrate, in ideal structures, in excitons with small 2D wave vectors $k_{ex} \leq k_{phot}$, appear to be distributed over a wider range of the lower excitonic states [8]. The function of distribution of the oscillator strengths $G(f)$ is a highly important characteristic of optical susceptibility and at the same time, as will be shown below, can characterize the quality of the SL-structure much more accurately and specifically then the HH-exciton's absorption bandwidth widely used for this purpose. The function $G(f)$ can be determined by any method which allows to measure evolution of population of optically active excitonic states generated under their resonance excitation with
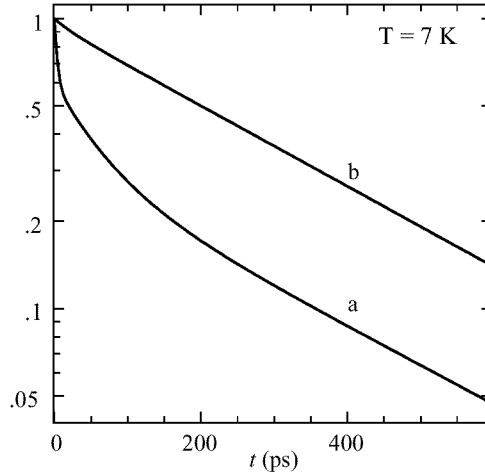
Fig 2.

no cascade-type population processes, which may involve rather long-lived intermediate states. In the method of nondegenerate differential photoreflection, this problem is solved by superposing the frequency of the pump fsec pulse onto the HH-exciton's absorption band maximum and by shifting the probe pulse frequency to long-wavelength side. For orthogonal polarizations of the pump and probe light pulses, the signal of the differential photoreflection is controlled by two processes with characteristic dependences on time delay between the pulses t_D. A part of the signal, we are interesting in, proportional to $\rho_{ex}(t)$, characterizes amplitude variation of coherent scattering of the probe beam due to the pump-induced changes of population of the excitonic states. The second process is related to non-scalar interaction controlling the coherent four-photon scattering.

Its dependence on t_D virtually coincides with the correlation function squared $K_{12}(t_D)$ of the pump and probe fields. Both signals can be easily extracted from experimental curve $\delta R(t_D)$ using the functional equality: $\rho_{ex}(t) \propto \delta R(t_D) - \text{const}|K_{12}(t_D)|^2$. The dependences $\rho_{ex}(t)$ obtained by this way for two SL (the nominaly pure SL_1-(a) and Fe-doped SL_2-(b)) and its multi-exponential fits:

$$\rho(t) \propto \sum_{i=1}^{3} A_i \exp(-\Gamma_1^i t) \qquad (1)$$

are shown in Fig. 2.

Where $A_1 = 0.41, A_2 = 0.32, A_3 = 0.27$ for SL_1 and $A_1 = 0, A_2 = 0.05, A_3 = 0.95$ for SL_2, $\Gamma_1^1 = 2.6 \cdot 10^{11}, \Gamma_1^2 = 1.7 \cdot 10^{10}, \Gamma_1^3 = 3 \cdot 10^9$ sec^{-1}. We will interpret these results based on the simplest model of 2D excitons in systems with structural disorder and associate the lowest rate observed in the experiment ($\Gamma_{1min} = 3 \cdot 10^9$ sec^{-1}) to radiative rate of the localized excitons with coherence radius $R_{coh} \approx a_B = 12$ nm. In this model, the ratio of the rates $\Gamma_{1i}/\Gamma_{1min}$ is determined by the ratio of areas of optical coherence of the corresponding states: $(R_{coh}/a_B)^2$. Thus the higest rate observed in experiment $\Gamma_{1max} = 2.6 \cdot 10^{11}$ sec^{-1} should be associated with excitons

having $R_{\mathrm{coh}} = 9a_{\mathrm{B}}$. The weights A_i evidently specify relative amount of the structure's sites which form the states with a given Γ_1^i.

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